Comment on "Spin Polarization and Magnetic Circular Dichroism in Photoemission from the 2*p* Core Level of Ferromagnetic Ni"

Recently, Menchero [1] applied the four-sites cluster model [2] to the interpretation of the $2p$ spin-resolved x-ray photoemission spectra (SRXPS) in Ni [3]. In this Comment we show, by applying the Ni_4 cluster to the $L_{2,3}$ magnetic circular dichroism (MCD), that the Ni ground state is not well described by this model and that it cannot provide a satisfactory description of all magnetic dichroic experiments.

The excitation of the core electron into the valence shell makes MCD more sensitive to ground-state properties than SRXPS, where the difference between the majority and minority spectrum is a result of final-state interactions between the core hole and the polarized valence shell. The calculation of the MCD spectrum, including a finite valence spin-orbit coupling (Fig. 1, dotted line), directly shows two major discrepancies between theory and experiment. First, the integrated intensities at the two spin-orbit split edges do not correspond to the experimentally observed ones. From the relations of these intensities to ground state expectation values of L_z and S_z [4,5] we find $\langle L_z \rangle / \langle S_z \rangle = 0.35$. This should be compared with the experimental value of $\langle L_z \rangle / \langle S_z \rangle = 0.19$ [5,6]. It is clear that the Ni_4 cluster overestimates the orbital magnetic moment. Second, the satellite structures, of mainly $d^8 \rightarrow \underline{p}d^9$ character, are absent.

These discrepancies are a direct result of the choice of the Ni_4 cluster. This model has a ground state consisting mainly of two holes of X_5 (t_{2g}-like) symmetry. This preference for one particular k point leads to a ground state that overestimates the orbital moment and has extremely small d^8 character, since the two holes avoid each other very effectively [2]. Choosing a two-hole ground state of X_2^2 (e_g-like, which is the second lowest state) or mixed X_2, X_5 character (by switching the energy ordering of the X_2 and X_5 band states) lowers the orbital moment to some extent, but still leads to a small d^8 character ($<3\%$).

We were able to obtain only a ground state with a relatively small $\langle L_z \rangle / \langle S_z \rangle$ of 0.24 and a significant d^8 character (12%) by having three holes of predominantly X_2 character. Unfortunately, the good agreement for SRXPS is then lost, as Fig. 1 (solid line) shows.

In conclusion, although the Ni_4 cluster includes more information regarding the Ni band structure with respect to the Anderson impurity model [7,8], it also favors very peculiar ground states which are incompatible with a coherent picture of all dichroism experiments. Any attempts to improve the situation by increasing the cluster size would imply formidable computational efforts.

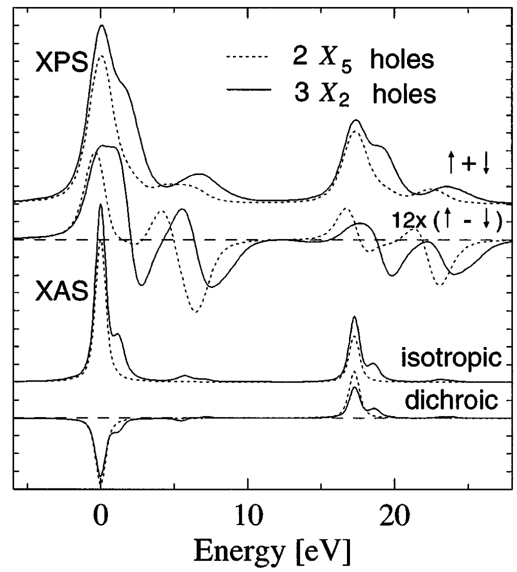

FIG. 1. The lower part shows the isotropic and circular dichroic $L_{2,3}$ x-ray absorption spectra (XAS) and the upper part gives the sum and the difference of the minority (↑) and majority (↓) $2p$ XPS spectra, in the same geometry as in Ref. [1]. Calculations are done for the Ni_4 cluster with two X_5 (dotted) and three X_2 (solid) holes.

Nicola Manini, Michel van Veenendaal,
and Massimo Altarelli
European Synchrotron Radiation Facility
B.P. 220
F-38043 Grenoble Cédex, France

Received 15 May 1997 [S0031-9007(97)04138-0]
PACS numbers: 75.25.+z, 75.30.Et, 78.20.Ls, 79.60.Bm

[1] J.G. Menchero, Phys. Rev. Lett. **76**, 3208 (1996); Phys. Rev. B **55**, 5505 (1997).
[2] L.M. Falicov and R.H. Victoria, Phys. Rev. B **30**, 1695 (1984); R.H. Victoria and L.M. Falicov, Phys. Rev. Lett. **55**, 1140 (1985).
[3] A.K. See and L.E. Klebanoff, Phys. Rev. B **51**, 11 002 (1995).
[4] B.T. Thole, P. Carra, F. Sette, and G. van der Laan, Phys. Rev. Lett. **68**, 1943 (1992).
[5] P. Carra, B.T. Thole, M. Altarelli, and X. Wang, Phys. Rev. Lett. **70**, 694 (1993).
[6] C.T. Chen, F. Sette, Y. Ma, and S. Modesti, Phys. Rev. B **42**, 7262 (1990); J. Vogel and M. Sacchi, Phys. Rev. B **49**, 3230 (1994).
[7] T. Jo and G.A. Sawatzky, Phys. Rev. B **43**, 8771 (1991).
[8] G. van der Laan and B.T. Thole, J. Phys. Condens. Matter **4**, 4181 (1992).

